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SERIAL NO. 10/092,128

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of )  
BARBEE *et al.* ) Confirmation No.: 9405  
Serial No.: 10/092,128 ) Art Unit: Unassigned  
Filed: March 06, 2002 ) Examiner: Unassigned  
For: "PROCESS FOR PREPARING HIGH )  
BARRIER NANOCOMPOSITES" )

PRELIMINARY AMENDMENT

Commissioner for Patents  
Washington, D.C. 20231

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May 14, 2002

Sir:

Prior to examination on the merits, please amend the captioned patent application as follows:

Refund Ref: 05/31/2002

0030007892

IN THE CLAIMS

Credit Card Refund Total: \$66.00

Please enter new claims 46-97 into this application:

Am Exp.: XXXXXXXXXXXX1008

46. A nanocomposite composition comprising about 0.05 weight percent to about 40 weight percent of a layered silicate material intercalated with an onium ion spacing agent and about 60 weight percent to about 99.95 weight percent of a matrix polymer comprising the reaction product of meta-xylylene diamine and a dicarboxylic acid, wherein the intercalated layered silicate material is dispersed uniformly throughout the matrix

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02 FC:103 18.00 GP

polymer.

47. A nanocomposite composition in accordance with claim 46, wherein the matrix polymer is co-intercalated into the layered silicate material.
48. A nanocomposite composition in accordance with claim 47, wherein the matrix polymer is co-intercalated into the layered silicate material while dispersing the layered material throughout the matrix polymer.
49. A nanocomposite composition in accordance with claim 47, wherein the matrix polymer is co-intercalated into the layered silicate material prior to dispersing the layered silicate material throughout the matrix polymer.
50. A nanocomposite composition in accordance with claim 46, wherein the matrix polymer is a polymer or oligomer of the reaction product of meta-xylylene diamine and adipic acid.
51. A nanocomposite composition in accordance with claim 46, wherein the onium ions include at least one moiety covalently bonded to a positively charged nitrogen atom that has a length of at least six carbon atoms.
52. A nanocomposite composition comprising a matrix polymer in an amount of about 40% to about 99.95% by weight, and about 0.05% to about 60% by weight of an intercalated phyllosilicate material formed by contacting a phyllosilicate with intercalant onium ions to form an intercalating composition, having a molar ratio of onium ions:phyllosilicate interlayer exchangeable cations of at least about 0.25:1 to achieve sorption of the onium ions between adjacent spaced layers of the phyllosilicate to expand the spacing between a predominance of the adjacent phyllosilicate platelets at least about 3 .ANG., when

measured after sorption of the onium ions, and a second intercalant disposed between adjacent spaced layers of the phyllosilicate material, said second intercalant comprising a nylon polymer formed by polymerizing a reaction product of meta-xylylene diamine and a dicarboxylic acid.

53. A composition in accordance with claim 52, wherein the intercalated phyllosilicate is exfoliated into a predominance of individual platelets.
54. A composition in accordance with claim 52, wherein the dicarboxylic acid is adipic acid.
55. A composition in accordance with claim 54, wherein the second intercalant is intercalated into the phyllosilicate from an intercalating composition containing said second intercalant in a concentration of at least about 5% by weight, based on the dry weight of the phyllosilicate in the intercalating composition.
56. A composition in accordance with claim 56, wherein the concentration of the second intercalant in said intercalating composition is at least about 20% by weight, based on the dry weight of the phyllosilicate in the intercalating composition.
57. A composition in accordance with claim 56, wherein the concentration of the second intercalant in said intercalating composition is at least about 30% by weight, based on the dry weight of the phyllosilicate in the intercalating composition.
58. A composition in accordance with claim 57, wherein the concentration of the second intercalant in said intercalating composition in the range of about 50% to about 80% by weight, based on the dry weight of the phyllosilicate in the intercalating compound.
59. A composition in accordance with claim 57, wherein the concentration of the second

intercalant in said intercalating composition in the range of about 50% to about 200% by weight, based on the dry weight of the phyllosilicate in the intercalating composition.

60. A composition in accordance with claim 52, wherein the molar ratio of intercalant onium ions:phyllosilicate interlayer exchangeable cations is at least 0.5:1.
61. A composition in accordance with claim 52, wherein the molar ratio of intercalant onium ions:phyllosilicate interlayer exchangeable cations is at least 1:1.
62. A composition in accordance with claim 52, wherein the onium ions are multi-onium ion compounds that include at least two primary, secondary, tertiary or quaternary ammonium, phosphonium, sulfonium, or oxonium ions.
63. A composition in accordance with claim 52, wherein the matrix polymer is MXD6 nylon.
64. A nanocomposite concentrate composition comprising about 10% by weight to about 90% by weight of a layered material intercalated with a polymer or oligomer of the reaction product of meta-xylylene diamine and a dicarboxylic acid and about 10 weight percent to about 90 weight percent of a matrix polymer comprising the reaction product of meta-xylylene diamine and a dicarboxylic acid, wherein the intercalated layered silicate material is dispersed uniformly throughout the matrix polymer.
65. A nanocomposite composition in accordance with claim 64, wherein the matrix polymer is intercalated into the layered silicate material.
66. A nanocomposite composition in accordance with claim 65, wherein the matrix polymer is intercalated into the layered silicate material while dispersing the layered material throughout the matrix polymer.

67. A nanocomposite composition in accordance with claim 65, wherein the matrix polymer is intercalated into the layered silicate material prior to dispersing the layered silicate material throughout the matrix polymer.
68. A nanocomposite composition in accordance with claim 64, wherein both the matrix polymer and the polymer intercalated into the layered material are a polymer or oligomer of the reaction product of meta-xylylene diamine and adipic acid.
69. A nanocomposite composition in accordance with claim 64, wherein prior to intercalating the layered material with the polymer of meta-xylylene diamine and a dicarboxylic acid, the layered material is first intercalated with onium ions that include at least one moiety covalently bonded to a positively charged nitrogen atom that has a length of at least six carbon atoms.
70. A method of decreasing oxygen permeability of a film or sheet of a matrix polymer comprising dispersing throughout said matrix polymer, in an amount of about 0.05% by weight to about 30% by weight, based on the total weight of the film or sheet material and the intercalate, an intercalate formed by intercalating an onium ion between layers of a layered silicate material, wherein said matrix polymer is a polymer or oligomer formed from the reaction product of a meta-xylylene diamine and a dicarboxylic acid, such that a portion of the matrix polymer is co-intercalated between the silicate layers of the layered material.
71. A method in accordance with claim 70, wherein the matrix polymer is an oxygen scavenger.
72. A method in accordance with claim 70, wherein the matrix polymer is co-intercalated into

the layered silicate material while dispersing the layered material throughout the matrix polymer.

73. A method in accordance with claim 70, wherein the matrix polymer is co-intercalated into the layered silicate material prior to dispersing the layered silicate material throughout the matrix polymer.
74. A method in accordance with claim 70, wherein the matrix polymer is a polymer or oligomer of the reaction product of meta-xylylene diamine and adipic acid.
75. A method in accordance with claim 70, wherein the onium ions include at least one moiety covalently bonded to a positively nitrogen atom that has a length of at least six carbon atoms.
76. A method of manufacturing a composite material containing about 10% to about 99.95% by weight of a matrix polymer comprising a polymer or oligomer of a reaction product of meta-xylylene diamine and a dicarboxylic acid, and about 0.05% to about 60% by weight of an intercalated phyllosilicate material, said intercalated phyllosilicate having an intercalant onium ion spacing agent intercalated between and bonded, by ion-exchange, to an inner surface of the phyllosilicate platelets, comprising: contacting the phyllosilicate with said intercalant onium ion spacing agent, to achieve intercalation of said intercalant onium ion spacing agent between said adjacent phyllosilicate platelets in an amount sufficient to space said adjacent phyllosilicate platelets a distance of at least about 3 .ANG.; and dispersing the intercalate throughout said matrix polymer to achieve intercalation of a portion of the matrix polymer between the phyllosilicate platelets.
77. The method of claim 76, wherein said phyllosilicate is contacted with said intercalant onium ion spacing agent, said phyllosilicate, and a nylon oligomer or polymer intercalant

formed from the reaction product of meta-xylylene diamine and a dicarboxylic acid, wherein the concentration of the onium ion spacing agent is in a molar ratio of onium ions:phyllosilicate interlayer exchangeable cations of at least 0.25:1.

78. The method of claim 77, wherein said phyllosilicate is contacted with said intercalant onium ion spacing agent, said phyllosilicate, and a nylon oligomer or polymer intercalant formed from the reaction product of meta-xylylene diamine and a dicarboxylic acid, wherein the concentration of the onium ion spacing agent is in a molar ratio of onium ions:phyllosilicate interlayer exchangeable cations of at least 0.5:1.
79. The method of claim 78, wherein said phyllosilicate is contacted with said intercalant onium ion spacing agent, said phyllosilicate, and a nylon oligomer or polymer intercalant formed from the reaction product of meta-xylylene diamine and a dicarboxylic acid, wherein the concentration of the onium ion spacing agent is in a molar ratio of onium ions:phyllosilicate interlayer exchangeable cations of at least 1:1.
80. A method of manufacturing a composite material containing about 40% to about 99.95% by weight of a matrix thermoplastic or thermosetting polymer, and about 0.05% to about 60% by weight of an intercalated phyllosilicate material, said intercalated phyllosilicate having an intercalant onium ion spacing/coupling agent intercalated between adjacent phyllosilicate platelets comprising: contacting the phyllosilicate with an intercalating composition including an intercalant onium ion spacing/coupling agent in a molar ratio of onium ions:phyllosilicate interlayer cations of at least 0.25:1, and a nylon oligomer or polymer intercalant formed from the reaction product of meta-xylylene diamine and a dicarboxylic acid, to achieve intercalation of said intercalant onium ion spacing/coupling agent and said nylon intercalant between said adjacent phyllosilicate platelets in an amount sufficient to space said adjacent phyllosilicate platelets at least an additional 3 .ANG.; combining the intercalated phyllosilicate with said thermoplastic or thermosetting

matrix polymer, and heating the matrix polymer sufficiently to provide for flow of said matrix polymer; and dispersing said intercalated phyllosilicate throughout said matrix polymer.

81. A method in accordance with claim 80, wherein the intercalating composition includes about 10% to about 200% by weight of said nylon intercalant, based on the dry weight of phyllosilicate contacted by said intercalating composition.
82. A method in accordance with claim 80, wherein the amount of onium ion spacing/coupling agent intercalated into the phyllosilicate material is in a molar ratio of at least 0.5:1, onium ions:exchangeable cations in the interlayer spaces of the phyllosilicate material.
83. A method in accordance with claim 82, wherein the amount of intercalant onium ion spacing/coupling agent intercalated into the phyllosilicate material is in a molar ratio of at least 1:1, onium ions:exchangeable cations in the interlayer spaces of the phyllosilicate material.
84. A method in accordance with claim 83, wherein the molar ratio of intercalated onium ion spacing/coupling agent to interlayer phyllosilicate cations is from about 1:1 to about 1:5.
85. A method in accordance with claim 80, wherein the weight ratio of the nylon intercalant to phyllosilicate material, dry basis is from about 20 grams of nylon intercalant per 100 grams of phyllosilicate material to about 200 grams of nylon intercalant per 100 grams of phyllosilicate material.
86. A method in accordance with claim 80, wherein the nylon oligomer or polymer is intercalated into the phyllosilicate by melting the nylon oligomer or polymer and

dispersing the phyllosilicate throughout the nylon melt.

87. A method in accordance with claim 86, wherein the mixing is accomplished in an extruder.
88. A method of manufacturing a composite material containing about 40% to about 99.95% by weight of an oligomer or polymer formed from the reaction product of meta-xylylene diamine and a dicarboxylic acid, and about 0.05% to about 60% by weight of an intercalated phyllosilicate material comprising intercalating the phyllosilicate material with an onium ion spacing agent by contacting the phyllosilicate with onium ions in a molar ratio of onium ions:phyllosilicate interlayer exchangeable cations of at least 0.25:1; forming a mixture of the intercalated phyllosilicate material with meta-xylylene diamine and a dicarboxylic acid; and subjecting the mixture to conditions sufficient to react and polymerize the meta-xylylene diamine with the dicarboxylic acid, to polymerize the meta-xylylene diamine and dicarboxylic acid reaction product while in contact with the intercalated phyllosilicate and to co-intercalate the resulting oligomer or polymer between adjacent platelets of the phyllosilicate material, wherein the intercalate reactants are combined in amounts such that the resulting composite material contains 40% to 99.95% oligomer or polymer and 0.05% to 60% intercalated phyllosilicate.
89. In a method of preventing the passage of oxygen to a material to be protected from oxygen contact comprising disposing a film or sheet of material between an oxygen source and the material to be protected, the improvement comprising the film or sheet material, said film or sheet material comprising a MXD6 nylon matrix polymer containing an intercalated phyllosilicate containing a MXD6 intercalant intercalated between adjacent phyllosilicate platelets in an amount sufficient to reduce the amount of oxygen contacting the material to be protected.

90. In the method of claim 89, wherein the amount of intercalate combined with the MXD6 nylon matrix polymer is in the range of about 2% to about 10% by weight intercalate, based on the total weight of the film or sheet material.

91. In the method of claim 89, wherein the amount of intercalate is in the range of about 3% to about 6% by weight.

92. In the method of claim 89, wherein the MXD6 intercalant is an oligomer formed from the reaction product of meta-xylylene diamine and adipic acid.

93. In the method of claim 92, wherein the amount of intercalate is at least about 4% by weight of the film or sheet material.

94. In the method of claim 93, wherein the amount of intercalate is at least about 5% by weight of the film or sheet material.

95. An intercalate formed by contacting a layered silicate material with an onium ion spacing/coupling agent intercalant, said intercalate having a molar ratio of intercalant onium ion spacing/coupling agent cation to interlayer cations of at least about 0.25:1, to achieve sorption and ion-exchange of the onium ion spacing/coupling agent with interlayer exchangeable cations of said layered silicate material to expand the spacing between a predominance of the adjacent platelets of said layered material at least about 3 .ANG., when measured after ion-exchange with the onium ion spacing/coupling agent; and a nylon oligomer or polymer second intercalant comprising the reaction product of meta-xylylene diainine and a dicarboxylic acid disposed between adjacent layers of said layered silicate material, to expand the spacing between a predominance of the adjacent platelets an additional at least 3 .ANG..

96. An intercalate in accordance with claim 95, wherein the layered silicate material is contacted with said onium ion spacing/coupling agent in an intercalant composition comprising said layered silicate material, said onium ion spacing/coupling agent and a carrier for said onium ion spacing/coupling agent and wherein the concentration of the onium ion spacing/coupling agent in said intercalating composition is at least about 2% by weight, based on the dry weight of the layered silicate material.

97. A method of intercalating a phyllosilicate with both an onium ion and an MXD6 nylon polymer comprising slurring the phyllosilicate in water to form a clay/water slurry; adding the onium ion to the phyllosilicate/water slurry; separating the onium ion-treated clay from the water; drying the phyllosilicate; and then mixing the onium ion-treated phyllosilicate with MXD6 nylon to intercalate the MXD6 nylon between adjacent platelets of the phyllosilicate.

**REMARKS**

This Preliminary Amendment and the new claims 46-97 submitted herewith are presented to satisfy the requirements of 37 C.F.R. §1.607(c). The new claims 46-97 correspond at least substantially to claims 1-52 of U.S. Patent No. 6,232,388 B1 to Lan *et al.*, issued May 15, 2001.

Credit Card Payment (Form PTO-2038) in the amount of \$1,776.00 is included to cover the \$996.00 fee under 37.C.F.R. §1.116(b) for 52 additional claims, and the \$840.00 fee for ten (10) additional independent claims under 37.C.F.R. §1.116(c). No other fees are believed due;

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however, the Commissioner is hereby authorized to charge any additional fees, which may be required, or credit any overpayment to Deposit Account No. 14-0629.

Respectfully submitted,

NEEDLE & ROSENBERG, P.C.

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I hereby certify that this correspondence and any documents referenced herein as being enclosed herein are being deposited with the United States Postal Service as Express Mail No. EL055858915US in an envelope addressed to: Commissioner for Patents, Washington, D.C. 20231 on the date below.

*Michael Moody*  
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Michael Moody

*5/14/02*  
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Date